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Spectral Function Analysis on Spin Dynamics in Double-Exchange Systems with Randomness

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(Received October 22,2002)

Spin excitation spectrum is studied in the double-exchange model with randomness. Applying the spin wave approximation and the spectral function analysis, we examine excitation energy and linewidth using analytical as well as numerical methods. For small wave number $q \sim 0$, the excitation energy is cosine-like and the linewidth shows a q-linear behavior. This indicates that the spin excitation becomes incoherent or localized near $q = 0$. Crossover takes place to marginally-coherent regime where both the excitation energy and the linewidth are proportional to q^2. The incoherence is due to local fluctuations of the kinetic energy of electrons. Comparison with experimental results in colossal magnetoresistance manganites suggests that spatially-correlated or mesoscopic-scale fluctuations are more important in real compounds than local or atomic-scale ones.

KEYWORDS: colossal magnetoresistance manganites, double-exchange model, randomness, spin wave excitation, linewidth

Introduction — One of main issues in colossal magnetoresistance (CMR) manganites $AMnO_3$ is whether the double-exchange (DE) mechanism is enough or not, and, if not, what is necessary as an additional mechanism.[1)] The metallic ferromagnetism is well described by the DE mechanism qualitatively,[2)] which indicates that the DE interaction is obviously an essential element in these materials. Recently, however, many experimental aspects have been indicated the necessity of additional elements beyond it. For instance, insulating behavior above the Curie temperature $T_{\rm C}$ as well as large residual resistivity is observed in compounds, such as (La,Ca)MnO_3, which show relatively low $T_{\rm C}$.[3)] The DE model predicts metallic conductivity both above and below $T_{\rm C}$ as observed in compounds which have high $T_{\rm C}$, such as (La,Sr)MnO_3.[4)] Thus, another mechanism might be necessary to describe the former 'low-$T_{\rm c}$' compounds. Since these deviations from the canonical DE behavior appear to be systematic for the A-site substitution,[5)] the question is what is the controlling parameter. The magnitude of CMR effects becomes larger in lower-$T_{\rm C}$ materials, therefore the issue attracts much attentions from the viewpoint of the basic mechanism of CMR.

One of crucial tests for an additional mechanism is spin dynamics. Spin excitation spectrum shows different behavior between high-$T_{\rm C}$ and low-$T_{\rm C}$ manganites. In high-$T_{\rm C}$ compounds, the spin excitation shows a cosine-like dispersion,[6)] which is well reproduced by the DE mechanism alone.[7)] On the contrary, in low-$T_{\rm C}$ compounds, the spectrum deviates from this form and exhibits some anomalies such as broadening, softening and gap-opening.[8,9,10,11)] Several mechanisms have been proposed to explain these anomalies.[12,14,13,15,16,17)] Most of them can reproduce qualitative features of the anomalous spin excitations to some extent, hence, more quantitative comparison between experimental and theoretical results is desired to determine an essential mechanism.

An intriguing point is that spin excitations in low-$T_{\rm C}$ manganites show large intrinsic linewidths even at the lowest temperature.[8,9,10,11,18)] This indicates that magnons in the ground state are not the eigenstates of the system. Theoretical models which can be effectively mapped to Heisenberg spin systems with uniform exchange couplings cannot describe this situation.

The authors have claimed that the anomalous spin excitation is well reproduced by introducing the randomness.[17)] Randomness appears to be promising among many proposed scenario since it is inherently controlled by the A-site substitution and gives a comprehensive understanding for systematic changes of the spin excitation from the high-$T_{\rm C}$ to the low-$T_{\rm C}$ materials, at least, in a qualitative level. In the presence of randomness, magnons are no longer the eigenstates, and the spin excitation shows a deviation from the cosine-like dispersion and a finite linewidth. It is strongly desired to compare the excitation spectrum including the linewidth quantitatively with experimental results.

In this Letter, we study the randomness effect on the spin wave spectrum quantitatively, especially on the excitation energy and the linewidth, within the spin wave approximation. We find an incoherent magnon excitation in the vicinity of the zone center and a crossover to a marginally-coherent regime where the linewidth is almost linearly scaled to the excitation energy.

Formulation — We investigate spin dynamics in the DE model with quenched randomness. The Hamiltonian includes both diagonal and off-diagonal disorder, which is

explicitly given by

$$\mathcal{H} = -\sum_{ij,\sigma} t_{ij}(c^\dagger_{i\sigma}c_{j\sigma}+\text{h.c.}) - J_{\rm H}\sum_i \boldsymbol{\sigma}_i\cdot\boldsymbol{S}_i + \sum_{i\sigma}\varepsilon_i c^\dagger_{i\sigma}c_{i\sigma}, \tag{1}$$

where the first term denotes the electron hopping, the second one is the Hund's-rule coupling between itinerant electrons and localized spins with a magnitude S, and the last one gives the on-site potential. The diagonal and off-diagonal disorder are incorporated in the on-site potential ε_i and the transfer integral t_{ij}, respectively. We call the former 'the on-site randomness' and the latter 'the bond randomness' hereafter. In the following, we first discuss these randomness effects on spin dynamics in a general formulation which does not depend on either details of the type of randomness or the dimension of the system. Later, we compare the analytic results with numerical ones.

By applying the spin wave approximation in the lowest order of $1/S$ expansion, the spin wave excitations in the limit of $J_{\rm H}/t \to \infty$ are obtained from the static part of the magnon self-energy,[17,7]

$$\Pi_{ij} = \frac{1}{2S}\sum_{mn} f_{n+}\varphi_{n+}(j)\varphi^*_{n+}(i)\varphi_{m-}(i)\varphi^*_{m-}(j)(E_m - E_n), \tag{2}$$

where f_{n+} is the fermi distribution function for up-spin states. Here $\varphi_{n\sigma}(i)$ is the n-th orthonormal eigenfunction, which satisfies $\sum_j \mathcal{H}_{ij}(\{t_{ij},\varepsilon_i\})\varphi_{n\sigma}(j) = (E_n - \sigma J_{\rm H})\varphi_{n\sigma}(i)$ for Hamiltonian (1) with a given configuration of randomness. The spectral function for the spin wave, $A(\boldsymbol{q},\omega)$, is calculated by averaging the quantity

$$A(\boldsymbol{q},\omega) = \frac{1}{N}\sum_l \Big|\sum_j \psi_l(j)\mathrm{e}^{\mathrm{i}\boldsymbol{q}\boldsymbol{r}_j}\Big|^2 \delta(\omega-\omega_l) \tag{3}$$

for random configurations. Here, N is the system size; ω_l and $\psi_l(j)$ are the eigenvalues and eigenfunctions of Π_{ij}, respectively, which satisfy

$$\sum_j \Pi_{ij}\psi_l(j) = \omega_l\psi_l(i). \tag{4}$$

Note that the observable quantities are to be averaged finally for configurations of randomness.

Before going into the analysis on the spectral function, we here discuss the magnon self-energy Π_{ij} in detail. Eq. (2) can be written in the form

$$\Pi_{ij} = \frac{1}{2S}\big(\mathcal{H}_{ij}B_{ji} - \delta_{ij}\sum_k \mathcal{H}_{ik}B_{ki}\big), \tag{5}$$

when we define $B_{ji} = \sum_n f_n\varphi_n(j)\varphi^*_n(i)$ and use the relations $\sum_m E_m\varphi_m(i)\varphi^*_m(j) = \sum_{mk}\mathcal{H}_{ik}\varphi_m(k)\varphi^*_m(j) = \mathcal{H}_{ij}$ and $\sum_n f_n E_n\varphi_n(j)\varphi^*_n(i) = \sum_k \mathcal{H}_{jk}B_{ki}$ with the orthonormal property of $\varphi_n(i)$. Here we drop the spin indices for simplicity. Apparently from eq. (5), Π_{ij} satisfies the sum rule

$$\sum_j \Pi_{ij} = 0. \tag{6}$$

Moreover, the matrix element Π_{ij} consists of the transfer energy of electrons as

$$\Pi_{i\neq j} = \frac{1}{2S}\mathcal{H}_{ij}B_{ji} = -\frac{1}{2S}t_{ij}\langle c^\dagger_i c_j\rangle \equiv -2SJ_{ij},$$
$$\Pi_{ii} = -\frac{1}{2S}\sum_{j\neq i}\mathcal{H}_{ij}B_{ji} = 2S\sum_j J_{ij}, \tag{7}$$

where $J_{ij} = t_{ij}\langle c^\dagger_i c_j\rangle/4S^2$ is the exchange coupling of the corresponding Heisenberg model within the lowest order of the spin wave expansion.[19] The bracket denotes the expectation value in the ground state for a given configuration of randomness. Hence the following summations equal to the kinetic energy of electrons as

$$\sum_i \Pi_{ii} = -\sum_{i\neq j}\Pi_{ij} = -\langle T\rangle/2S, \tag{8}$$

where T is the first term in the Hamiltonian (1).

Now we apply the spectral function analysis on the spin excitation spectrum. By using the m-th moment

$$\Omega^{(m)}_{\boldsymbol{q}} = \int_0^\infty \omega^m A(\boldsymbol{q},\omega)d\omega, \tag{9}$$

the excitation energy $\omega_{\rm sw}$ and linewidth γ of the spin wave excitation are obtained by

$$\omega_{\rm sw}(\boldsymbol{q}) = \Omega^{(1)}_{\boldsymbol{q}}, \tag{10}$$

$$\gamma^2(\boldsymbol{q}) = \Omega^{(2)}_{\boldsymbol{q}} - (\Omega^{(1)}_{\boldsymbol{q}})^2, \tag{11}$$

respectively. The moments in eq. (9) can be calculated by the magnon self-energy Π_{ij}. By using eqs. (3) and (4), we obtain

$$\Omega^{(m)}_{\boldsymbol{q}} = \frac{1}{N}\sum_{ij}\sum_{k_1k_2\ldots k_{m-1}}\Pi_{ik_1}\Pi_{k_1k_2}\cdots\Pi_{k_{m-1}j}\mathrm{e}^{\mathrm{i}\boldsymbol{q}(\boldsymbol{r}_i-\boldsymbol{r}_j)}. \tag{12}$$

Namely, $\Omega^{(m)}_{\boldsymbol{q}}$ is a Fourier transform of Π^m_{ij}. This spectral function analysis is valid only if the excitation spectrum is single-peaked. Previous study shows that this is the case as long as $\boldsymbol{q}\sim 0$ even in the presence of randomness.[17] This will be demonstrated also in Fig. 1 later. Thus, we can discuss the excitation energy and linewidth at $\boldsymbol{q}\sim 0$ by this spectral function analysis.

First, we analyze the excitation energy (10). Hereafter, we assume the electron hopping only for nearest-neighbor sites in model (1) for simplicity. Then, by eq. (5), Π_{ij} has nonzero matrix elements only for $i=j$ and $j+\boldsymbol{\eta}$. Here $\boldsymbol{\eta}$ is a displacement vector to the nearest neighbor site. From eqs. (10) and (12), the excitation energy is given by

$$\omega_{\rm sw}(\boldsymbol{q}) = \frac{1}{N}\sum_i\Big(\Pi_{ii} + \sum_{\boldsymbol{\eta}}\Pi_{i,i+\boldsymbol{\eta}}\,\mathrm{e}^{\mathrm{i}\boldsymbol{q}\boldsymbol{\eta}}\Big)$$
$$= \sum_{\boldsymbol{\eta}}\bar{\Pi}(\boldsymbol{\eta})(\mathrm{e}^{\mathrm{i}\boldsymbol{q}\boldsymbol{\eta}}-1), \tag{13}$$

where we define the site-averaged quantity $\bar{\Pi}(\boldsymbol{\eta}) = \frac{1}{N}\sum_i \Pi_{i,i+\boldsymbol{\eta}}$ and use the sum rule (6). The symmetry for the direction of $\boldsymbol{\eta}$ is expected to be recovered after the random average, therefore we assume $\bar{\Pi}(\boldsymbol{\eta}) \equiv -\Lambda$ to be irrespective of $\boldsymbol{\eta}$. Finally, the spin-wave excitation

energy is expressed as

$$\omega_{\rm sw}(\boldsymbol{q}) = \Lambda \sum_{\boldsymbol{\eta}} (1 - {\rm e}^{{\rm i}\boldsymbol{q}\boldsymbol{\eta}}). \tag{14}$$

Thus, for hypercubic lattices, the spectrum has the cosine form for small $\boldsymbol{q}$ even in the presence of disorder. The quantity Λ gives the spin stiffness since

$$\omega_{\rm sw}(\boldsymbol{q}) \simeq \Lambda q^2 \tag{15}$$

in the limit of $q = |\boldsymbol{q}| \to 0$. The spin stiffness is proportional to the kinetic energy of the electronic Hamiltonian (1) in our analysis as easily shown by eq. (8).

Next, we consider the linewidth (11). Similarly to eq. (13), the second moment is written as

$$\Omega^{(2)}_{\boldsymbol{q}} = \sum_{\boldsymbol{\eta}_1 \boldsymbol{\eta}_2} \bar{\Pi}_2(\boldsymbol{\eta}_1, \boldsymbol{\eta}_2)(1 - {\rm e}^{{\rm i}\boldsymbol{q}\boldsymbol{\eta}_1})(1 - {\rm e}^{{\rm i}\boldsymbol{q}\boldsymbol{\eta}_2}), \tag{16}$$

where $\bar{\Pi}_2(\boldsymbol{\eta}_1, \boldsymbol{\eta}_2) = \frac{1}{N}\sum_i \Pi_{i+\boldsymbol{\eta}_1, i}\Pi_{i, i+\boldsymbol{\eta}_2}$. In the absence of disorder, we have $\Pi_{i+\boldsymbol{\eta}, i} = \Pi_{i, i+\boldsymbol{\eta}} = -\Lambda$. Hence the linewidth γ becomes zero since $\Omega^{(2)}_{\boldsymbol{q}} = (\Omega^{(1)}_{\boldsymbol{q}})^2$. In the presence of disorder, the linewidth becomes finite. For simplicity, we consider a special $\boldsymbol{q}$ such as $(q, 0, 0, \cdots)$ on a hypercubic lattice. Denoting two different cases of $\boldsymbol{\eta}_1 = \boldsymbol{\eta}_2$ and $\boldsymbol{\eta}_1 = -\boldsymbol{\eta}_2$ as

$$\bar{\Pi}_2(\boldsymbol{\eta}_1, \boldsymbol{\eta}_2) = \Lambda_2 \pm \delta\Lambda_2 \quad \text{for } \boldsymbol{\eta}_1 = \pm\boldsymbol{\eta}_2, \tag{17}$$

we obtain

$$\Omega^{(2)}_{\boldsymbol{q}} = 4\big(\Lambda_2(1 - \cos q)^2 + \delta\Lambda_2 \sin^2 q\big). \tag{18}$$

In the limit of small $\boldsymbol{q}$, the linewidth is estimated as

$$\gamma^2(\boldsymbol{q}) \simeq 4\delta\Lambda_2 q^2 + \big(\Lambda_2 - \frac{4}{3}\delta\Lambda_2 - \Lambda^2\big)q^4. \tag{19}$$

Therefore, there is a q-linear contribution in the linewidth γ in the presence of disorder.

Consequently, we obtain $\omega_{\rm sw} \propto q^2$ and $\gamma \propto q$ in the limit of $\boldsymbol{q} \to 0$. This indicates that the spin wave excitation becomes incoherent or localized in the vicinity of $\boldsymbol{q} = 0$ since $\omega_{\rm sw} < \gamma$. This incoherent behavior comes from local fluctuations of the transfer energy of electrons since the coefficient of the $\boldsymbol{q}$-linear term in γ is proportional to

$$\delta\Lambda_2 \propto \bar{\Pi}_2(\boldsymbol{\eta}, \boldsymbol{\eta}) - \bar{\Pi}_2(\boldsymbol{\eta}, -\boldsymbol{\eta}) \propto \sum_i \big(\Pi_{i+\boldsymbol{\eta}, i} - \Pi_{i-\boldsymbol{\eta}, i}\big)^2$$

$$\propto \sum_i \big(J_{i+\boldsymbol{\eta}, i} - J_{i-\boldsymbol{\eta}, i}\big)^2. \tag{20}$$

As increasing $\boldsymbol{q}$, the incoherent regime is taken over by the marginally-coherent one where $\omega_{\rm sw} \propto \gamma \propto q^2$. We will discuss this crossover in comparison with numerical results in the following.

Results — For further understanding, we calculate the excitation energy and the linewidth numerically, and compare them with the above analytic expressions. In numerical calculations, we obtain the spectral function $A(\boldsymbol{q}, \omega)$ following the method in ref. 17, and calculate the moments by the definition (9). In the following, we discuss two-dimensional cases as an example. The qualitative feature of the spectrum is almost independent of the dimension of the system. The system size is 40×40 sites and the electron density is fixed at $n = \sum_i \langle c_i^\dagger c_j\rangle/N = 0.7$. The random average is taken for 50 different realizations of random configurations. We examine effects of the on-site and the bond randomness separately. In both cases, we consider the binary-alloy-type distribution of randomness as $\varepsilon_i = \pm\delta\varepsilon$ and $t_{ij} = t \pm \delta t$, where the sign takes plus or minus in equal probability. We note that other type distributions, such as Gaussian-type, do not change conclusions. As an energy unit, we use the half bandwidth $W = 1$ at the ground state for $J_{\rm H} = \delta\varepsilon = \delta t = 0$. The dimensionless constant $2S$ is set to be unity.

Figure 1 shows a typical excitation spectrum in the presence of randomness. The gray-scale contrast shows the intensity of the spectrum, namely, the magnitude of the spectral function $A(\boldsymbol{q}, \omega)$. The global structure keeps a portion of the cosine-like one in the case of pure system,[7] however, some anomalies appear near the zone boundaries. This is due to the Friedel oscillation of J_{ij} in eq. (7).[17] The gray curve shows the excitation energy $\omega_{\rm sw}(\boldsymbol{q})$ calculated from eq. (10). As clearly seen, $\omega_{\rm sw}(\boldsymbol{q})$ well describes the excitation spectrum in the small-$\boldsymbol{q}$ regime where the anomalies are not substantial. This strongly supports the applicability of the above spectral analysis with eqs. (9)-(11) in this regime.

The inset in Fig. 1 shows the numerical results for $\omega_{\rm sw}(\boldsymbol{q})$ and $\gamma(\boldsymbol{q})$ from $\boldsymbol{q} = (0, 0)$ to $(\pi, 0)$. They follow $\omega_{\rm sw} \propto q^2$ and $\gamma \propto q$ in the small-$\boldsymbol{q}$ region as predicted in eqs. (15) and (19). At $\boldsymbol{q} \sim 0$, there is the incoherent regime where $\gamma > \omega_{\rm sw}$ as discussed above.

In Fig. 2, we compare effects of the on-site and the bond randomness by plotting the linewidth as a function of the excitation energy. For the bond randomness, the behavior $\gamma \propto \omega_{\rm sw}^{1/2}$ dominates the spectrum as shown in the figure. On the contrary, for the on-site randomness, the $\omega_{\rm sw}^{1/2}$ part is observed only in the small-$\boldsymbol{q}$ region, and the marginally-coherent behavior $\gamma \propto \omega_{\rm sw}$ is dominant in the wide region of $\boldsymbol{q}$. Thus, the $\boldsymbol{q}$-linear contribution in the linewidth is more dominant in the case of the bond randomness compared to the case of the on-site one. This indicates that the bond randomness tends to make the spin excitation more incoherent than the on-site one.

As discussed above the $\gamma \propto \omega_{\rm sw}^{1/2}$ behavior is due to local fluctuations. Therefore, this different aspect between the on-site and the bond randomness indicates that the bond randomness induces larger local fluctuations than the on-site one. This is qualitatively understood as follows. There are two factors which contribute to the local fluctuations of the transfer energy $(J_{i+\boldsymbol{\eta}, i} - J_{i-\boldsymbol{\eta}, i})^2$ in eq. (20). One is the expectation value $\langle c_i^\dagger c_j\rangle$ and the other is the transfer integral t_{ij}. In the case of the on-site randomness, we have uniform $t_{ij} = t$, hence only $\langle c_i^\dagger c_j\rangle$ contributes to the local fluctuations. The expectation value $\langle c_i^\dagger c_j\rangle$ is disordered, however, it may have some correlations due to the Friedel oscillation. By the Friedel oscillation, the charge density shows a correlation whose length scale is $\sim 2k_{\rm F}$, where $k_{\rm F}$ is the Fermi wave number. On the contrary, in the case of the bond randomness, t_{ij} fluctuates from site to site independently in model (1). This may suppress the correlation in $\langle c_i^\dagger c_j\rangle$.

Therefore, we have stronger local fluctuations and more incoherent spin excitation in the case of the bond randomness than in the on-site one.

Comparison with experiments — In real materials, some spatially-correlated or mesoscopic-scale randomness may exist due to, for instances, A-site clustering or twin structure of lattices. Our results suggest that if these kinds of correlated randomness are substantial, the incoherent regime diminishes and the marginally-coherent behavior $\gamma \propto \omega_{\rm sw}$ dominates the spectrum since they may reduce local fluctuations. Experimentally, the incoherent behavior $\gamma \propto \omega_{\rm sw}^{1/2}$ has not been clearly observed yet. Perring *et al.* have reported an approximately linear scaling of $\gamma \propto \omega_{\rm sw}$ in the wide region of the spectrum in layered perovskite manganites.[18] They observed a significantly large damping, that is, $\gamma/\omega_{\rm sw} \simeq 0.3 - 0.5$. Our results in the marginally-coherent regime in Fig. 2 (a) are consistent with these experimental observations. Therefore, we speculate that the randomness plays an important role on the spin dynamics in CMR manganites, and that correlated or mesoscopic-scale randomness might be substantial compared to atomic-scale one.

Another possible origin for the linewidth broadening which survives down to the lowest temperature is the magnon-electron interaction.[15,16] In this scenario, the linewidth is predicted to be $\gamma \propto q^{d+3}$ where d is the dimension of the system. For $d = 2$ or 3, this q dependence is stronger than in our result (19), and is inconsistent with the above experimental result.

Summary — We have studied the spin dynamics in the double-exchange model with quenched randomness within the spin wave approximation in the lowest order of $1/S$ expansion. We have derived analytic expressions for the excitation energy and the linewidth by the spectral function analysis. Incoherent magnon excitation is found in the vicinity of the zone center where the linewidth is proportional to the square-root of the excitation energy. As increasing the wave number, this incoherent behavior is taken over by the marginally-coherent one in which the linewidth is proportional to the excitation energy. Atomic-scale randomness enhances the incoherence through the local fluctuations of the kinetic energy of electrons. Comparison with experimental results is satisfactory and suggests an importance of correlated or mesoscopic randomness in real materials.

This work is supported by "a Grant-in-Aid from the Mombukagakushou".

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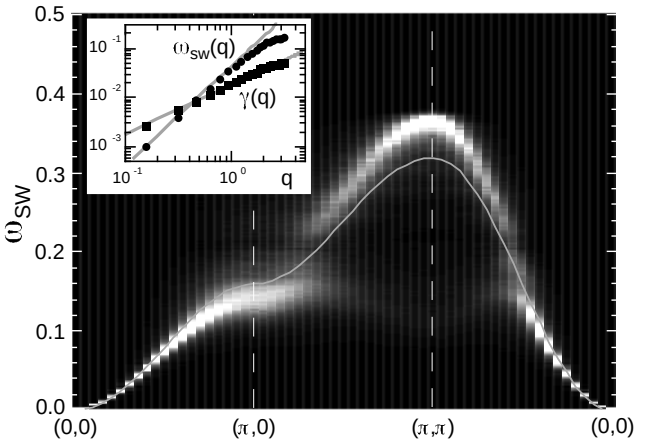

Fig. 1. Spin excitation spectrum in the case of the on-site randomness $\delta\varepsilon = 0.25$. The gray curve is the excitation energy calculated by eq. (10). Inset: The excitation energy $\omega_{\rm sw}$ and the linewidth γ as a function of $(q, 0)$ for $0 < q < \pi$. The lines are the fits by q^2 and q for $\omega_{\rm sw}$ and γ, respectively.

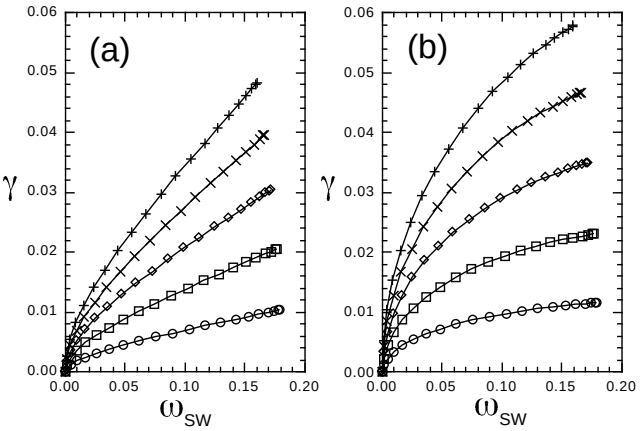

Fig. 2. The linewidth plotted as a function of the excitation energy in the case of (a) the on-site randomness $\delta\varepsilon = 0.05, 0.1, 0.15, 0.2, 0.25$ and (b) the bond randomness $\delta t = 0.025, 0.05, 0.075, 0.1, 0.125$ from bottom to top, respectively.